ANTHROPOGENIC ORGANIC CONTAMINANTS IN AMERICAN LOBSTERS (*Homarus americanus)* PROCURED FROM HARBOURS, BAYS, AND INLETS OF EASTERN CANADA

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ABSTRACT

Market-sized (0.80 ± 0.12 kg) lobsters were collected in 1995 from nineteen locations in eastern Canada including one Offshore. The Offshore location was selected as the control location for this study. ∑PAH concentrations ranged from <8 to 14000 $ng \cdot g^{-1}$ wet wt. Saint John Harbour, an area of vast industrial activity in the Bay of Fundy, surprisingly had one of the lowest concentrations of PAHs. The South Arm of Sydney Harbour, Nova Scotia, a closed fishery for lobster due to decades of coke-oven discharge contamination, and Dartmouth Cove of Halifax Harbour, contained the highest PAH concentrations in lobster digestive glands. Humber Arm, Newfoundland lobster contained PAH levels comparable to Halifax Harbour lobster. Benzo[*a*]pyrene was reported in lobster from all sites except the Offshore site. Sydney Harbour and Humber Arm lobsters contained the highest levels of benzo[*a*]pyrene. 4H-Cyclopenta[*def*]phenanthrene, a suspected carcinogen, found in lobster from most sites, should be analysed routinely with the common PAHs.

Chlorobiphenyls patterns and levels in lobster are site specific. The ∑CB concentrations ranged from 94 to 1900 $ng \cdot g^{-1}$ wet wt. The highest concentrations were found in Sydney Harbour, Halifax Harbour, East Dover, Humber Arm and Petit-de-Grat, which were the same sites, except East Dover, that contained the highest PAH results. The offshore control site contained surprising levels of CBs. CBs 118/106. 138/163, 153, and 180 are the major bioaccumulators in lobster from all locations sampled. These are persistent organochlorine compounds found in environmental samples worldwide. Lobster from Petit-de-Grat contains the highest levels of CB 118, a mono-*ortho* CB, which is toxic. Toxic equivalent 2,3,7,8-tetrachlorodibenzo-*p*-dioxin concentrations for this CB in Petit-de-Grat lobster slightly exceeds Health Canada's tolerance for 2,3,7,8-tetrachlorodibenzo-*p*-dioxin. Several of the Halifax Harbour lobster contained CBs, which are not present in the common Aroclor mixtures. IUPAC nos. 11 and 108 were reported in an earlier study on seals from Sable Island and was detected in lobster from Halifax Harbour.

RÉSUMÉ

En 1995, des homards de taille commerciale (0,80 ± 0,12 kg) ont été échantillonnés à 19 endroits dans l'est du Canada, dont un endroit au large qui a servi de site témoin pour cette étude. Les homards ont présenté des concentrations de ∑HAP qui variaient de <8 à 14 000 $ng \cdot g^{-1}$ poids humide. Curieusement, une des concentrations de HAP les plus basses a été observée dans le port de Saint-Jean, un secteur d'intense activité industrielle dans la baie de Fundy. Les concentrations de HAP les plus élevées ont été trouvées dans les glandes digestives de homards capturés dans le bras sud du port de Sydney (Nouvelle-Écosse), où la pêche du homard est fermée en raison de décennies de contamination par les effluents de fours de cokerie, et dans l'anse Dartmouth du port de Halifax. Les homards du bras Humber (Terre-Neuve) présentaient des niveaux de HAP comparables à ceux du port de Halifax. Du benzo[*a*]pyrene a été détecté dans les homards de tous les sites sauf celui situé au large, et les homards du port de Sydney et du bras Humber

présentaient les plus grandes concentrations de ce composé. En plus des HAP courants, il faudrait mesurer de façon routinière la concentration de 4H-cyclopenta[def]phénanthrène, un carcinogène présumé trouvé dans les homards de la plupart des sites.

Les concentrations absolues et relatives des chlorobiphényles (CB) dans les homards varient selon le site. Les concentrations de $\sum$CB ont varié de 94 à 1 900 $ng \cdot g^{-1}$ poids humide et elles étaient les plus élevées dans les homards des ports de Sydney et de Halifax, du bras Humber, de Petit-de-Grat et d'East Dover, soit les mêmes sites, à l'exception du dernier, où les teneurs en HAP étaient les plus élevées. Les concentrations de CB étaient étonnamment élevées dans le site témoin au large. Les CB 118/106, 138/163, 153 et 180 ont présenté la plus forte bioaccumulation dans les homards de tous les sites échantillonnés. Il s'agit de composés organochlorés persistants présents dans des échantillons environnementaux partout au monde. Les homards de Petit-de-Grat contenaient les plus fortes concentrations de CB 118 (un CB mono-*ortho* toxique), lesquelles correspondaient à une toxicité équivalente à des concentrations de 2,3,7,8-tétrachlorodibenzo-*p*-dioxine supérieures à la norme de Santé Canada. Plusieurs des homards échantillonnés dans le port de Halifax contenaient des CB absents des mélanges d'Aroclor courants. Détectés chez des phoques de l'île de Sable dans une étude antérieure, les composés n° 11 et 108 selon la nomenclature de l'UICPA étaient présents dans les homards du port de Halifax.

1.0 INTRODUCTION

One of the most valuable fisheries in eastern Canada is the commercial lobster fishery, which exists mainly in the near-shore area, including many commercial harbours, bays, and inlets. The heavily industrialised harbours, Sydney, Halifax, and Saint John, yield the largest commercial catches of all harbours in the area (Prouse 1994). Chlorobiphenyls (CBs) do not occur naturally, but rather were produced commercially by the direct chlorination of biphenyl. Two hundred and nine congeners are produced by this process and these compounds are non-flammable and relatively insoluble in water. These properities made CBs very desirable for a variety of industrial applications including hydraulic fluids, solvent extenders, transfer fluids, flame retardants, carriers of inks and dyes, and plasticizers in paints and adhesives (Nimii and Oliver 1989). In North American most of these commercial mixtures were prepared and marketed by Monsanto under the brand name Aroclors. Polycyclic aromatic hydrocarbons (PAHs) are present in the environment due to both natural and anthropogenic sources. The two main sources of PAHs in the environment are pyrogenic, combustion of fossil fuels and vegetation, and petrogenic, petroleum products, inputs. PAHs and polychlorinated biphenyls (PCBs) are widespread contaminants of the marine environment. PAHs and PCBs are lipophilic compounds that tend to accumulate in the fatty organs of fish. For the past two decades there has been concern over the toxic effects of these compounds on man and marine life.

Elevated PAH concentrations in the digestive gland (hepatopancreas) of American lobster (*Homarus americanus*) from the South Arm of Sydney Harbour, Nova Scotia resulted in the closure of the lobster fishery in 1982. Lobsters from Sydney Harbour were re-examined in 1992, and a number of additional PAHs and heterocylcic aromatic hydrocarbons were identified (King et al., 1993). High PAH concentrations, although well below those recorded in Sydney Harbour, have also been found in lobsters from Halifax Harbour, Nova Scotia (Uthe et al., 1989; Prouse, 1991). Although many of the sites in this study have been analysed in the past for PCBs, very few of the sites have been analysed for CB congeners if at all, due to the unavailability of individual chlorobiphenyls standards and the use of packed-column gas chromatography which is inadequate for resolving PCB mixtures (Uthe et al. 1988). Early techniques for measuring PAHs and PCBs used HPLC flourscence and gas chromatograpthy with electron capturing detection respectively. These techniques are more susceptible to interference from compounds that are similar chemically and come through the extraction and cleanup procedures in tact and interfer with measurement of PAHs and PCBs. This study used high resolution gas chromatography coupled to mass spectrometry (GC-MS), a highly sophisticated analytical instrument, allowing accurate and precise measurements of organic contaminants. We focused on lobster from 19 commercially fished sites covering four provinces, Nova Scotia, New Brunswick, Newfoundland, and Quebec, to fill in missing background contaminant data and to assess the marine environmental quality for the region.

2.0 MATERIALS AND METHODS

2.1 Sampling

Market-size (0.80 ± 0.12 kg) lobsters were collected using standard commercial fishing gear in 1995 from nineteen locations in eastern Canada; South Arm of Sydney Harbour, Nova Scotia (immediate area of coal tar discharge and untreated sewage discharge); Dartmouth Cove (industrial inputs, area of urban runoff and untreated sewage discharge), McNabs (island in the central part of the harbour, Point Pleasant (cargo containment area and municipal park), Portuguese Shore, and Trumpcap of Halifax Harbour, Petit-de-Grat Harbour, (area of fish plant waste discharge), East Dover, Liverpool Harbour (fish processing plant and industrial activity), Minas Basin, Pictou Harbour (industrial inputs), St. Margarets Bay, Nova Scotia; Belledune Harbour (high industrial inputs), Dalhousie Harbour (industrial inputs), and Saint John Harbour (industrial inputs and immediate area of ocean dumping), New Brunswick; Humber Arm, Cornerbrook Newfoundland (area of high industrial inputs); Magdalen Islands (in the Gulf of Lawrence approximately 120 kms from Prince Edward Island), and Grand Rivière, Québec; and Georges Bank (offshore control site) (Figure 1).

2.2 Sample Preparation

Lobsters were transported live to the laboratory. The digestive glands (pooled from 10 animals) were removed weighed and frozen (-20°C) prior to sample preparation for chemical analyses. Digestive glands were thawed and homogenised using a Polytron tissue homogenizer (Model PT 10 20 3500), weighed (2.00g) into mixing cylinders and PAH and PCB surrogates internal standards were added. Next the tissues were saponified in 3.6 M aqueous ethanolic potassium hydroxide for 1.5 hrs and extracted into hexane (Musial and Uthe 1986, King et al., 1993 and 1996). The extracts were purified on a gel-permeation chromatograph (Autoprep, Analytical BIO Chemistry Laboratories, Model 1001 GPC) fitted with a jacketed 4 cm i.d. glass column (maintained at 19 °C and packed with EnvioBeads, S-X3 select ATS Scientific Inc., 200-400 mesh, 30 cm column bed length) and a Schoeffel Monochromator GM 770 (used at 254 nm). After GPC cleanup the extracts were divided in half, one half for acid treatment prior to CB analysis and the second for PAH analysis.

2.3 *Chemical Analyses*

The acid treated portion of the extracts were analysed for CBs (159 compounds; IUPAC Nos. 1-16, 18-22, 24-32, 34-40, 42, 44, 47-50, 52-55, 58, 60-62, 65-66, 69-70, 72, 75, 77-82, 86-88, 93, 95, 98-106, 108-110, 112, 114-119, 121-124, 126-129, 131-134, 136-145, 147, 149, 151-161, 163, 165-171, 173, 180-192, 194-196, 199-202, and 204-209 Ultra Scientific Canada) on a Hewlett Packard Model 5890 Series II gas chromatograph interfaced to a Model 5971 mass selective detector (electron ionization) and fitted with a cool on-column inlet and a chromatographic column (30m x 0.25 mm id. fused silica coated with SPB-5, film thickness 0.25µm, Supelco, Canada).

The second portion of the extract was analysed for PAHs (Ultra Scientific Canada and National Institute of Standards and Technology, SRM 1492) on a Hewlett-Packard gas chromatograph with a MS-Engine (Hewlett Packard 5989B-electron and negative chemical ionization modes) fitted with a (pressure pulse) split/splitless inlet and chromatograhic column (30m x 0.25mm i.d. fused silica coated with HP-5MS, film thickness 0.25µm,

Hewlett Packard, Canada). Both GC-MS systems were used in the selective ion monitoring (SIM) modes.

2.4 Fortifications Studies

Fortification studies (2.0, 10.0, and 100.0 ng·g-1 wet wt added to tissue) yielded recoveries that ranged 63.1 to 104% for all CBs [mean recovery ± standard deviation = 88.8 ± 8.9 with relative standard deviations (n= 3 to 5) of 6.2-21.4% for CB classes ranging from mono- to decachlorobiphenyl (Table 1). As expected, the recoveries became more erratic as the amount of spike decreased. Recoveries of ^{13}C-CBs, IUPAC Nos. 153 and 194 ranged from 75 to 105% [mean recovery ± relative standard deviation = 88 ± 11 %] and 76 to 106% [mean recovery ± relative standard deviation = 91 ± 10%] respectively. Recoveries of d_{10}-anthracene, d_{12}-benz[*a*]anthracene, d_{12}-benzo[*b*] fluoranthene, d_{12}-benzo[*a*]pyrene, and d_{14}-dibenz[*a,h*]anthracene were 78-105% [mean recovery ± relative standard deviation = 90 ± 8%], 77-104% [mean recovery ± relative standard deviation = 89 ± 9%], 87-112% [mean recovery ± relative standard deviation = 101 ± 6%], 89-103% [mean recovery ± relative standard deviation = 98 ± 3%], and 85-110% [mean recovery ± relative standard deviation = 100 ± 7%], respectively. Due to the poor recoveries of d_8-naphthalene, results were not reported for naphthalene.

SRM IAEA-142 was anlysed with each batch of 10 samples. Results for the standard reference material were valid within 1 to 2 standard deviation from the mean certified values. Blanks, SRM, and duplicate samples were processed with each batch of 10 samples.

3.0 RESULTS AND DISCUSSION

3.1 Polycyclic Aromatic Hydrocarbons and Polychlorinated Biphenyls

$\sum$PAH concentrations (Tables 2-3) ranged from <8 for Georges Bank to 14,000 ng·g^{-1} wet wt. for South Arm of Sydney Harbour (14,000 ng·g^{-1} wet wt.), followed by Halifax Harbour (Dartmouth and Point Pleasant Park), Humber Arm of Newfoundland, all sites with large populations, major industrial sources and/or a history of organic contamination problems. Surprisingly, Belledune, Saint John, and Dalhousie Harbours of New Brunswick had relatively lower PAH concentrations in lobster digestive glands compared to Nova Scotia harbours i.e., Halifax Harbour. These harbours receive major industrial and raw sewage discharge. Magdalen Islands of Québec had relatively low PAH concentrations in lobster digestive gland compared to Nova Scotia. Grande Rivière had lower PAH levels than the Magdalen Islands. However, Georges Bank an offshore site had, as expected, undetectable PAHs in lobster digestive gland.

Chlorobiphenyls patterns and levels in lobster are site specific (Tables 5-6). The summed PCB concentrations ranged from 94 to 1900 ng·g^{-1} wet wt. The highest concentrations were found in Sydney Harbour, Halifax Harbour, East Dover, and Petit-de-Grat, which were the same sites that had the highest PAH concentrations, except East Dover, that contained the highest PAH results. Petit-de-Grat Harbour contains a fish process plant, which discharges its waste into the harbour. The effluents and process streams from the

fish meal plant contain high PCB concentrations (0.83 $mg \cdot L^{-1}$) and this waste discharge is believed to be the source of PCB contamination in the harbour (Ernest et al. 1982). This would also explain the high ΣCB concentration found in lobster from this location. A total of 101 CBs have been identified in lobster collected in Nova Scotia, with lesser numbers and lower concentrations of CBs in lobsters from other provinces. CBs show smaller ranges of concentrations compared to PAHs, but are detected at all sites. IUPAC Nos. 118/106, 138/163, 153, and 180 with averages ± standard deviations of 52.7±5.7% (Nova Scotia excluding East Dover and Portuguese Shore, but including the offshore site), 55% (Newfoundland one site), 41.7±4.0% (New Brunswick), and 47.5% (Québec average of two sites) are the major CBs congeners bioaccumulated by lobsters from all four provinces. The consistent uptake of these congeners by lobster with small variation between the site locations suggests that lobsters are a good bioindicator for monitoring CBs in the marine environment

3.1.1 Nova Scotia and Offshore

A large range of PAH concentrations were observed in this study with the highest concentrations restricted to the inner parts of harbours with known PAH sources. Most of the lobster fishing occurs in the inner to outer reaches of Halifax Harbour. Approximately 65 fishing boats occupy the harbour and fish 7200 traps in the outer harbour. There are four lobster-holding facilities located in the harbour. The value of the fishery exceed one million Canadian dollars annually (Prouse 1994). PAH levels in lobster decrease rather rapidly as distance increased away from the source location. Lobsters collected in Dartmouth Cove, in the inner harbour very close to one of the major outfalls and a great deal of shipping activity, and Point Pleasant Park areas, within about 2 kilometres of the major municipal and industrial sewers, oil refineries and other sources of PAHs, have the second and third highest PAH concentrations observed in our study 2200 and 1300 $ng \cdot g^{-1}$, respectively. Samples collected near McNabs Island (4 kms from the sources - 650 $ng \cdot g^{-1}$), Thrumcap Shoal (10 kms - 480 $ng \cdot g^{-1}$) and Portuguese Shore (12 kms -300 $ng \cdot g^{-1}$) show rapidly decreasing concentrations with distance from the inner harbour sources. Measuring PAHs in sediment samples in the locations where lobsters were collected may help to identify some of the sources for PAHs that were present in lobster digestive glands. Since the lobster were collected in harbour and bays that are near municipal and industrial discharge there may be a number of potential sources for PAHs. An earlier study indicated that sediments collected in Halifax Harbour contained elevated levels of PAHs ranging from 510 to 25,000 $ng \cdot g^{-1}$ dry wt. (Tay et al. 1992). Our results confirm that commercially fished lobsters, from many industrial harbours, accumulate high levels of PAHs, including common ones thought to be carcinogenic. This is of great concern, because lobster, even after transported and held in a clean environment for one year, do not lose substantial amounts of all PAHs (Uthe and Musial 1986).

The individual CB patterns vary considerably from location to location. The Sydney sample is perhaps the most distinctive. Ninety eight percent of the congeners in this sample are penta- to nonachlorobiphenyls, very much an Aroclor 1260 pattern. The samples from Portuguese Shore and East Dover are more than 50% di- to tetrachorobiphenyl, with major peaks that are indicative of a predominantly Aroclor 1242/1254 mixture. The samples collected at the four sites; Dartmouth Cove, Point

Pleasant Park, McNabs Island, and Trumpcap in Halifax Harbour are on average 86.7±5.7% penta- to heptachlorobiphenyl suggesting an Aroclor 1254/ 1260 mixture.

The offshore control site contained surprising high levels of CBs. We expected the levels of CBs from this site to be much lower near detection limits. Since this site is not near any industrial activity, the higher than expected values suggests that the lobsters could have recently migrated to the area or due to the persistent nature of these compounds lobster maybe accumulating selective congeners over time. PAHs were non-detectable in lobster from this area.

3.1.2 Newfoundland

Only one site was selected from Newfoundland based on industrial inputs. The Bay of Islands, located on the West Coast of the Island, is a large body of water measuring in total approximately 355 km^2 (137 sq. MI). It is composed of four main parts: an open bay, dotted with about twelve islands, for which the bay was named, and three large arms which extend from the open water eastward: North Arm, Middle Arm (which itself divides into two smaller extensions of the sea: Goose and Penguin Arms) and the Humber Arm, into which the Humber River empties. Glacial scouring of the area caused the waters of the arms and bay to be quite deep with steep banks. Salmon fishing attracted settlers to the shores of the Bay of Islands. Development of herring fishing, saw mills and a large pulp and paper industry in Corner Brook in made the eastern end of the Humber Arm the area with the second largest population in Newfoundland after St. John's. Humber Arm lobster contained PAH levels comparable to Halifax Harbour lobster. The only difference between these two locations was that Halifax Harbour lobster contained alkylated PAHs, but these compounds appear to be present in the inner most reaches of the harbour. The Humber River is located on the westcoast of Newfoundland and flows into the Humber Arm of the Bay of Islands. Corner Brook is located at the mouth of the Humber River. This highly industrial area explains the high ∑PAH results in lobster of 1200 $ng \cdot g^{-1}$ wet wt. The ∑CB concentration was 820 $ng \cdot g^{-1}$ wet wt. and this was also comparable to lobster collected from the outer reaches of Halifax Harbour.

3.1.3 New Brunswick

Dalhousie Harbour has a number of industrial activities, including an electric power plant, pulp and paper mill, chlor-alkali plant, and ore-loading facilities. Petroleum residues in sediments were 5-28 $mg \cdot kg^{-1}$ collected in the area in 1985 (Levy et al.). Organics were measured in lobster collected from Eel Bay (Matheson and Bradshaw (1985). The PCBs were near background and PAHs were not measured. Despite the industrial activity near this site, the summed PAH and PCB concentrations calculated in this study were low (210 and 290 $ng \cdot g^{-1}$ wet wt. respectively) confirming the earlier work on PCB in lobster by Matheson.

Lobsters from several other major industrial harbours, notably Saint John Harbour, are much less contaminated. Based on the PAH results presented here, Saint John Harbour is one of the least contaminated areas, not one of the worst as predicted in Prouse (1994).

The Saint John Harbour sampling location was, in fact, the dredgedump site, approximately 8 km from presumed inner harbour sources of PAHs. A decrease in concentration is reported for Halifax Harbour with distance from the inner harbour sources, a similar trend was reported for Sydney Nova Scotia (Prouse et al. 1994). If the same were true for Saint John Harbour this could explain the low concentrations found near the dredge spoil dumpsite. This would imply that the dredge spoils are not themselves a significant source of PAHs. Alternatively it could be that the lobsters caught near the dredge site were recent migrants to the area. As a result of the above findings and public concerns about contamination at the dumpsite further work on lobster from the Saint John Harbour area is warranted. PAH results in lobster from other sites were quite consistent with the predictions in the Prouse report (1994).

Belledune is a rapidly growing industrialised area in New Brunswick. Belledune is home to many industries such as a large lead/zinc smelter, a port (dry and liquid bulk deep water facility), batter and fertilizer plant, hydroelectric plant, high capacity saw mill, shell bulk plant (storage depot beside the port), and a Canadian gypsum processing plant. Despite all this industrial activity the PAH and PCB concentrations in lobster were low from this area, but lobster from this location contained slightly higher levels of PAH and PCB compared to the other sites selected in New Brunswick.

3.1.4 Québec

The Magdalen Islands are located in the Gulf of St. Lawrence, 120 km from Prince Edward Island. Between 1970, when the Irving Whale sank, and 1990, over 1100 tons of oil (27 per cent of its original cargo) have been released into the Gulf of St. Lawrence (Gilbert and Walsh 1996). Initially the Magdalen Islands were the most severely impacted area when 200 tons of oil washed ashore at the time of the sinking, and small amounts of oil have occasionally washed ashore over the past 25 years since then. As a result of this environmental disaster marine life has become contaminated with PAHs from the crude oil and PCBs from leakage of the heat-transfer unit. Lobster procured from the Magdalen Islands contained $\sum$PAH concentration of 750 $ng \cdot g^{-1}$ wet wt. the fifth highest in the study and it is most probable that the source of PAHs were from the oil release by the Irving Whale barge during the time of sinking. Also, Magdalen lobster contained alkylated PAHs. Despite the fact that the oil from Irving Whale washed ashore on the Magdalen Islands the PCB concentrations in lobster from this area were low.

Grande-Rivière got its name because of the river that goes through it. The harbour is a lively place with its numerous pleasure, sport and commercial fishing boats. Grande Rivière lobster contained one of the lowest $\sum$PAH concentrations. The same was the case for PCB concentrations in lobster from this area.

3.1.5 Contaminants of Concern Found in Lobster

The polynuclear aromatic hydrocarbons selected for analysis include both low molecular weight (1-3 rings) and high molecular weight (4-6 rings) compounds. A number of these are suspected carcinogens (Dong et al., 1978 and Lee et al., 1976) including some less

commonly analysed ones. For example, 4H-Cyclopenta[*def*]phenanthrene has been identified at a number of the sites. 4H-Cyclopenta[*def*] phenanthrene, a suspected carcinogen, found in lobster from most sites, should be analysed routinely with the common PAHs. Benzo[a]pyrene (carcinogen according to the World Health Organisation) ws reported for all locations, except the offshore control site. The World Health Organisation has calculated toxic equivalent factors for selected PAHs (dibenz[*a,h*]anthracene, benz[*a*]anthracene, benzo[*b*]fluoranthene, benzo[*k*]fluoranthene, chrysene, benzo[*ghi*]perylene, and indeno[1,2,3-*cd*]pyrene). These factors can be applied to the selected PAH concentrations to calculate a toxic equivalent bnzo[*a*]pyrene concentration. Since most of these toxic PAHs were found in lobster from our sampling locations it is valid to state that appling these factors to the selected PAHs would increase the benzo[*a*]pyrene concentrations in lobster. Benzo[a]pyrene concentrations in pooled lobster digestive glands ranged 8 for East Dover and Minas Basin to 790 $ng \cdot g^{-1}$ wet wt for Sydney Harbour. South Arm of Sydney Harbour lobster digestive glands contained Benzo[*a*]pyrene at 790 $ng \cdot g^{-1}$ wet wt., this concentration was very similar to 840 $ng \cdot g^{-1}$ wet wt. found in lobster by Sirota et al. (1983) 15 years ago from the same site. Humber Arm Newfoundland contained 730 $ng \cdot g^{-1}$ benzo[*a*]pyrene ranking second to Sydney Harbour. Halifax was third with 60 $ng \cdot g^{-1}$ benzo[*a*]pyrene found in lobster from Dartmouth Cove. Pictou Harbour was very close to Halifax with a benzo[*a*]pyrene concentration of 59 $ng \cdot g^{-1}$.

CB pair 118/106 is a major contributor to the summed PCB concentrations. If our source of PCBs comes from Aroclors then 106 can be removed from the pair, since it is not present in Aroclors (Frame et al. 1996). CB 118 is a mono-*ortho* substituted CB and has a toxic equivalence factor to 2,3,7,8-tetrachlorodibenzo-*p*-dioxin of 0.0001 (Ahlborge et al. 1994). If we apply this factor to the concentration of CB 118, the highest of all sites, found in lobster from Petit-de-Grat, we get a toxic equivalence concentration of 21 $pg \cdot g^{-1}$ wet wt. This value slightly exceeds the 20 $ng \cdot kg^{-1}$ wet wt. Canadian tolerance for 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (Health Canada 1993). Therefore, CB 118 would be a major contributor to the toxic equivalent concentrations calculated for lobster. Addison et al (1999) reported CB 118 to be the major constituent found in grey seals from Sable Island, Nova Scotia. Toxic equivalence concentrations were not calculated in this study. The above example was used to show the impact that CB 118, as a major contributor, would have on the toxic equivalence calculations.

Several of the samples also contain CBs (IUPAC Nos. 11 and 108) that are not present (<0.01 wt%) in the common Aroclor mixtures (Schultz et al. 1989 and Frame et al. 1996). IUPAC no. 11 was a significant peak in the Point Pleasant Shoal (Halifax Harbour) sample, and IUPAC no. 108 was found in most of the samples. IUPAC no. 108 was reported in seals from Sable Island (Addison et al. 1999). It appears odd that this compound is present, since it is not found in Aroclors. Addison gave no explanation for its presence in grey seals from Sable Island. We know from recent work that there is a source of low molecular wt. PCBs (mostly CB 11) from pigments found in ink production in the Burnside area near Tuff's Cove of Halifax Harbour (King et al. 2002). Examinating the possibility that CB 108 could be the result of dechlorination of PCBs in sediments was not apparent from our review of the literature (Bedard and May 1996; Brown and Wagner 1989). Neither study reported CB 108 as a dechlorination product.

Our standard mix of CBs does not contain all the pentachlorobiphenyls; therefore 108 could have a possible co-eluter this appears to be the only explanation. IUPAC no. 11 was also found in mussels from a nearby Halifax Harbour site (King et. al., inpress), otherwise we know of only two other cases where this CB was reported at trace levels in fish from Hudson River (Bush et al., 1989) and $pg \cdot g^{-1}$ (lipid wt) in grey seals from Sable Island, Nova Scotia (Addison et al., 1999). CB no. 11 is a member of the more toxic non-ortho class of CBs and this important from a toxicity standpoint. If has been reported that CB 11 produces neurochemical effects in rat cerebellar granule cells (Kodavanti 1995).

4.0 CONCLUSIONS

The study provides background information for work on future time-series studies. Areas such Sydney, Halifax, and Petit-de-Grat, Nova Scotia and Humber Arm Newfoundland, which contained some of the highest organic contaminant levels in lobster digestive gland warrants further investigation through monitoring programs.

The present work shows the importance of studying individual compounds by indentifying new contaminant sources, ie, CB no. 11 from pigments in ink production. This demonstrates that not all CBs found in the environment come from Aroclor mixtures.

The study clearly shows the ability of lobster to accumulate high levels of PAHs and PCBs in their digestive glands. In particular with the CBs the most dominant ones in all cases were IUPAC nos. 118/106, 138/163, 153, and 180.

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TABLES

Table 1. Mean percentage recoveries and RSDs of added chlorinated biphenyl to biological tissue.

Class	Fortification\ ng.g^{-1} wet wt.	mean	SD	Mean recovery (%)	SD	RSD (%)
mono	2.00	0.03	0.01	63.1	13.5	21.4
	10.00	0.15	0.02	73.2	10.5	14.3
	100.0	1.53	0.19	76.7	9.7	12.6
di	2.00	0.04	0.01	71.8	14.7	20.5
	10.00	0.18	0.03	88.0	15.9	18.1
	100.0	1.78	0.19	88.8	9.6	10.8
tri	2.00	0.04	0.01	81.3	16.2	19.9
	10.00	0.18	0.02	87.8	8.3	9.5
	100.0	1.84	0.16	91.8	8.1	8.8
tetra	2.00	0.04	0.01	89.8	15.6	17.4
	10.00	0.18	0.02	90.2	9.7	10.8
	100.0	1.85	0.14	92.6	7.1	7.7
penta	2.00	0.04	0.01	82.6	13.6	16.5
	10.00	0.20	0.02	98.2	10.2	10.4
	100.0	1.85	0.12	92.4	6.2	6.7
hexa	2.00	0.04	0.01	87	15.3	17.6
	10.00	0.20	0.03	99.2	17.2	17.3
	100.0	1.92	0.12	95.8	5.9	6.2
hepta	2.00	0.04	0.01	85.6	14.8	17.3
	10.00	0.21	0.03	104.0	14.0	13.5
	100.0	1.98	0.20	98.8	10.1	10.2
octa	2.00	0.04	0.01	89.0	15.6	17.5
	10.00	0.19	0.02	94.8	10.6	11.2
	100.0	1.95	0.15	97.4	7.5	7.7
nano	2.00	0.04	0.01	87.4	13.5	15.4
	10.00	0.19	0.01	93.2	7.0	7.5
	100.0	1.93	0.13	96.6	6.4	6.6
deca	2.00	0.04	0.01	82.6	15.7	19
	10.00	0.18	0.01	89.0	5.8	6.5
	100.0	1.90	0.14	95.0	6.8	7.2
Grand mean				88.8		10.1

Table 2. PAHs $ng \cdot g^{-1}$ wet wt. in digestive gland pools of lobster collected in the Halifax Harbour and St. Margarets Bay of Nova Scotia. A blank space indicates that the compound was not found at our detection limit (DL) of $<8\ ng \cdot g^{-1}$.

	Halifax Harbour					control
Compound	Dartmouth Cove	Point Pleasant Park	McNabs	Trumpcap	Portuguese Shore	St. Margarets Bay
2,7-dimethylnaphthalene	15	40				
acenaphthene	14	25	10			17
fluorene	26	34	14	12	11	20
1-methylfluorene	15	32	8			
phenanthrene	170	155	44	36	29	79
anthracene	31	15	15	14	18	19
4H-cyclopenta[*def*] phenanthrene	25	36	13	9		17
3,6-dimethylphenanthrene	69	27				
fluoranthene	590	361	170	97	75	330
pyrene	460	180	130	99	44	140
benz[*a*]anthracene	56	32	21	21	18	47
chrysene/triphenylene	380	145	94	72	47	140
benzo[*a*]fluorene	59	57	21	18	11	
benzo[*b*]fluorene	16	28	8	8		
benzo[*b*+*k*]fluoranthene	140	83	38	43	26	52
benzo[*a*]pyrene	60	26	26	25	13	23
indeno[123,*cd*]pyrene	57	32	18	12		13
benzo[*ghi*]perylene		18	12		8	
dibenz[*a*,*c*+*a*,*h*]anthracene	17		12	10		
∑PAH	2200	1300	650	480	300	890

Table 3. PAHs $ng \cdot g^{-1}$ wet wt. in digestive gland pools of lobster collected from other sites in Nova Scotia. A blank space indicates that a compound was not found at our DL of <8 $ng \cdot g^{-1}$ wet wt.

Compound	East Dover	Liverpool	Minas Basin	Pictou Harbour	Petit-De-Grat	South Arm	Georges Bank control
acenaphthylene						28	
acenaphthene	10	24	16		24	31	
fluorene	17	24	16	15	31	100	
phenanthrene	57	42	41	31	200	440	
anthracene	8	8			15	130	
4H-cyclopenta[*def*] phenanthrene					22	120	
3,6-dimethylphenanthrene	8				20		
fluoranthene	80	58	41	23	370	3900	
pyrene	39	34	20	9	160	2700	
benz[*a*]anthracene					22	790	
chrysene/triphenylene	17	16	31	8	84	1700	
benzo[*a*]fluorene					22	490	
benzo[*b*]fluorene					9	160	
benzo[*b+k*]fluoranthene	16	16	15	100	33	1600	
benzo[*a*]pyrene	8	10	8	59	13	790	
indeno[123,*cd*]pyrene						47	
benzo[*ghi*]perylene						520	
dibenz[*a,c+a,h*]anthracene	9					61	
ΣPAH	270	230	190	250	1000	14000	

Table 4. PAHs $ng \cdot g^{-1}$ wet wt. in digestive gland pools of lobster collected from Newfoundland (Nfld), New Brunswick, and Québec Canada. A blank space indicates that the compound was not found at our DL of $<8\ ng \cdot g^{-1}$ wet wt.

	Nfld	New Brunswick			Québec	
Compound	Humber Arm	Belledune	Saint John	Dalhousie	Magdalen Islands	Grand Rivière
2,7-dimethylnaphthalene					11	
acenaphthene	8	14			91	9
2,3,5-trimethylnaphthalene					15	
fluorene	11	14		13	49	14
1-methylfluorene					9	
phenanthrene	76	59	24	43	120	28
anthracene	15	11	8		11	
4H-cyclopenta[*def*] phenanthrene	15	9			17	
3,6-dimethylphenanthrene	18					
fluoranthene	350	98	47	62	220	46
pyrene	240	85	21	35	93	29
benz[*a*]anthracene	59	12			16	
chrysene/triphenylene	150	45	23	26	45	24
benzo[*a*]fluorene	37	13			13	
benzo[*b*]fluorene	14					
benzo[*b*+*k*]fluoranthene	120	21	17	17	25	20
benzo[*a*]pyrene	73	10	13	9	15	11
indeno[123,*cd*]pyrene	41					
dibenz[*a*,*c*+*a*,*h*]anthracene	12					
∑PAH	1200	400	150	210	750	180

Table 5. CBs $ng \cdot g^{-1}$ wet wt. in pooled digestive gland of lobster collected in Halifax Harbour and St. Margarets Bay of Nova Scotia. *- A peak was detected, but not all confirmation ions were satisfied. A blank space indicates that the CB was not detected at our DL <2.0 $ng \cdot g^{-1}$ wet wt.

		Halifax Harbour					control
		Dartmouth Cove	Point Pleasant Park	McNabs	Trumpcap	Portuguese Shore	St. Margarets Bay
IUPAC No.	Class	DCNS	PtPNS	MNNS	TCNS	PSNS	StMBNS
4/10	di		*			2.7	
8/5	di	*	*		*	7.7	
11	di	*	20	3.3	*		
15	di		*		*	2.9	
19	tri						
18	tri	3.5	7.4		*	27	
27/24	tri		*		*	2.0	
16	tri	*	6.3		*	21	
26	tri				*	4.1	
28/31	tri	16	21	6.9	10	61	3.5
21/33/20	tri	6.4	8.7		7.6	20	
22	tri	*	4.5		2.9	11	
36	tri		*		*		
37	tri	4.9	4.4		3.4	7.9	
53	tetra		*			8.5	
52	tetra	11	17	2.8	6.0	53	
49	tetra	6.2	*		4.4	33	
47/48/75	tetra	*	12	4.0	8.4	21	
44	tetra	7.1	15		4.0	47	
42	tetra	3.8	5.8		*	19	
40	tetra	2.4	*		*	9.9	
61/74	tetra	25	19	11	12	34	2.9
70	tetra	14	24	5.6	4.4	71	2.1
66/80	tetra	34	26	14	13	50	3.2
60	tetra	15	*	4.9	*	43	
77	tetra	4.9	3.5		*	4.0	
102/98	penta	*	*			2.5	
95/93	penta	7.4	8.8	2.9	5.9	20	
101	penta	25	20	12	11	27	2.2
99	penta	57	35	32	24	27	5.5
119	penta	2.9	2.1		*		
97/86	penta	4.1	4.8		8.5	11	
117/87/116/115	penta	14	11	5.0	*	14	
110	penta	33	28	13	14	47	3.1
82	penta	2.5	3.3		*	6.8	
124	penta	2.3	2.1		*		
108	penta	8.2	5.9	3.9	4.0	3.7	
123	penta	4.2	*		2.6		
118/106	penta	180	95	99	63	65	13
114	penta	5.1	3.1	2.5	*	2.5	
105/127	penta	66	28	28	24	28	3.9
126	penta	*	*	2.0	*		

Table 5 Continued

151	hexa	4.8	4.1	2.6	3.9	2.1	
144	hexa	*	*	2.6	*	2.5	
147	hexa	*	*		*		
149/139	hexa	21	17	13	15	9.4	
133	hexa	*	*	3.0	*		
153	hexa	230	170	190	130	69	21
132/168	hexa		*	3.7	*	3.8	
141	hexa	5.7	5.0	3.4	*	2.3	
137	hexa	14	7.3	6.5	5.8	3.3	
138/163	hexa	210	130	130	80	50	15
160/158	hexa	36	16	18	17	6.9	2.7
159	hexa	*	*	2.0	*		
128/167	hexa		29	30		13	3.4
156	hexa	22	11	13	*	5.7	
157	hexa	3.9	2.3	2.2	2.7		
187/182	hepta	69	72	75	50	25	7.4
183	hepta	18	16	14	12	5.2	
181	hepta	*	5.6	3.2			
171	hepta	9.6	*	7.3	*	2.4	
192	hepta	*	*	5.1	5.9		
180	hepta	100	97	110	77	30	7.6
191	hepta	2.8	2.1				
170/190	hepta	37	33	36	28	10	2.6
189	hepta	2.7	*		*		
202	octa	4.9	*	4.6	*	2.2	
200/201	octa	*	*		*		
196	octa	16	20	18		7.8	2.0
195	octa	4.2	5.0	4.2	*		
194	octa	13	14	14	*	4.6	
208	nona	*	2.2		2.4		
206	nona	3.9	4.6	2.4	*		
ΣCB		1400	1090	950	650	1070	100

Table 6. CBs $ng \cdot g^{-1}$ wet wt. in pooled digestive gland of lobster collected from other fishing sites in Nova Scotia. *- A peak was detected, but not all confirmation ions were satisfied. A blank space indicates that the CB was not detected at our detection limit <2.0 $ng \cdot g^{-1}$ wet wt.

IUPAC No.	Class	East Dover	Liverpool	Minas Basin	Pictou Harbour	Petit-De-Grat	South Arm	Georges Bank control
4/10	di	3.7		*				
6	di	2.0						
8/5	di	10	3.0	*		*	*	
15	di	3.9					*	
19	tri	2.4						
18	tri	38						
27/24	tri	3.2						
16	tri	28						
26	tri	5.1						
28/31	tri	76	4.5	4.8	6.4	9.6	7.0	4.6
21/33/20	tri	26	*	*	2.3	*	*	
22	tri	14						
37	tri	9.0				2.5	2.0	
53	tetra	11						
52	tetra	68	*	2.9	5.2	6.1	3.8	2.8
49	tetra	43		2.1	2.5	*	*	
47/48/75	tetra	27	4.7	4.4	2.9	9.1	*	2.7
44	tetra	59		*	3.5	3.3	*	
42	tetra	23						
40	tetra	11						
61/74	tetra	36	4.3	4.0	5.4	25	7.5	4.9
70	tetra	86	3.9	4.4	6.7	9.0	4.7	4.2
66/80	tetra	54	5.7	5.1	7.5	25	9.4	6.1
60	tetra	50	*	*	4.2	9.5	*	2.8
77	tetra	4.0				3.2	2.0	
102/98	penta	2.8						
95/93	penta	23	*	*	2.9	5.3	5.9	
101	penta	28	5.4	3.2	7.7	18	14	3.9
99	penta	20	7.6	5.1	10	66	18	12
119	penta					2.7		
97/86	penta	12		2.4		*	2.7	
117/87/116/115	penta	15	*	*	3.4	12	*	*
110	penta	49	6.5	4.2	11	24	13	5.1
82	penta	7.7						
108	penta	2.6	*			10	3.3	
123	penta					4.0		
118/106	penta	37	16	9.9	23	210	46	27
114	penta					6.2	*	
105/127	penta	20	5.6	*	8.1	60	2.8	9.1
126	penta		*			*	*	
151	hexa		2.6			2.7	*	
144	hexa		*			*	*	
149/139	hexa	5.8	7.9	3.1	6.1	11	36	3.1
133	hexa					5.6	*	

Table 6 Continued

153	hexa	22	42	16	32	240	390	55
132/168	hexa	3.1	*	*	2.3			
141	hexa	2.0	3.8			4.3	11	
137	hexa		*			11	*	
138/163	hexa	17	33	10	25	180	250	35
160/158	hexa	2.8	4.3		3.2	22	32	
159	hexa		*			*	*	
128/167	hexa	4.8	*	*	6.4		31	8.1
156	hexa	2.0	*		2.6	19	14	3.9
157	hexa					3.5	*	
187/182	hepta	7.6	27	7.2	11	48	250	18
183	hepta		7.3		2.0	11	54	3.0
181	hepta		6.0			*	15	
171	hepta		*			5.2	24	
192	hepta		3.1			4.1	18	
180	hepta	7.9	40	5.6	9.4	69	340	19
191	hepta					*	3.5	
170/190	hepta	2.4	*	*	4.7	20	100	6.8
189	hepta					*	4.8	
202	octa		*	*		3.3	15	*
200/201	octa		*	*		*	9.1	
196	octa	2.0	11	*		3.7	59	*
195	octa		*			*	16	
194	octa		8.9	*	*	6.5	46	*
208	nona						*	
206	nona		3.1			*	6.4	
ΣCB		990	270	94	220	1200	1900	240

Table 7. CBs $ng \cdot g^{-1}$ wet wt. in pooled digestive gland of lobster collected from fishing sites in New Brunswick, Québec, Newfoundland, and Offshore (control site). *-A peak was detected, but not all confirmation ions were satisfied. A blank space indicates that the CB was not detected at our detection limit. 2.0 $ng \cdot g^{-1}$ wet wt.

		Nfld.	New Brunswick			Québec	
IUPAC No.	Class	Humber Arm	Belledune	Saint John	Dalhousie	Magdalen Islands	Grande Rivère
28/31	tri	10	8.4	13	6.1	3.4	6.6
21/33/20	tri	4.8	*	7.3	*		*
37	tri	2.5		2.9			
52	tetra	5.7	6.3	8.9	5.1	2.0	5.2
49	tetra	3.0	*	5.7	*		2.8
47/48/75	tetra	8.6	5.8	*	6.0	2.3	*
44	tetra	3.7	3.3	8.0	2.6		*
42	tetra	*		3.6			
61/74	tetra	18	13	11	8.0	4.6	8.9
70	tetra	9.3	9.2	13	7.2	2.5	6.8
66/80	tetra	20	15	15	10	4.8	12
60	tetra	9.9	7.7	8.6	4.7		4.6
77	tetra	*		2.0			
95/93	penta	4.5	3.6	5.3	6.1		
101	penta	12	10	5.5	10	7.1	9.5
99	penta	26	20	14	16	12	15
97/86	penta	3.2		4.1	3.2		2.8
117/87/116/115	penta	9.1	4.6	8.0	7.5	2.8	7.4
110	penta	15	14	7.9	15	7.4	12
82	penta			2.2			
108	penta	4.4	2.8	3.0	3.1	2.1	2.9
123	penta	2.1					
118/106	penta	89	42	30	30	31	29
114	penta	3.4		*			*
105/127	penta	23	15	11	11	8.7	11
151	hexa	*			2.3		2.2
144	hexa	*	2.2		2.8		*
149/139	hexa	*	7.4	*	9.9	5.4	8.3
133	hexa	*	2.4	*			*
153	hexa	150	71	52	39	57	38
132/168	hexa	*	2.4	*	*		*
141	hexa	4.7			2.5		2.4
137	hexa	6.2	2.4	3.4	*		2.7
138/163	hexa	120	47	36	34	39	31
160/158	hexa	18	6.2	4.3	3.8	4.6	*
128/167	hexa	27	10	12		8.3	9.9
156	hexa	13	3.8	*	3.0	2.9	2.4
187/182	hepta	47	26	19	14	19	15
183	hepta	13	5.0	3.9	3.3	3.5	3.5
181	hepta	3.1					
171	hepta	6.6	2.3	*	*		
192	hepta	*		*	2.0		
180	hepta	90	27	19	15	10	15

TABLE 7 CONTINUED

170/190	hepta	*	7.6	9.2	6.5	3.5	*
202	octa	3.8	2.1	*	*		*
196	octa	15	4.7	5.3	*		4.0
195	octa	3.9		*			*
194	octa	*	3.4	4.5			*
206	nona	3.4		*			
∑CB		820	410	360	290	250	280

FIGURES

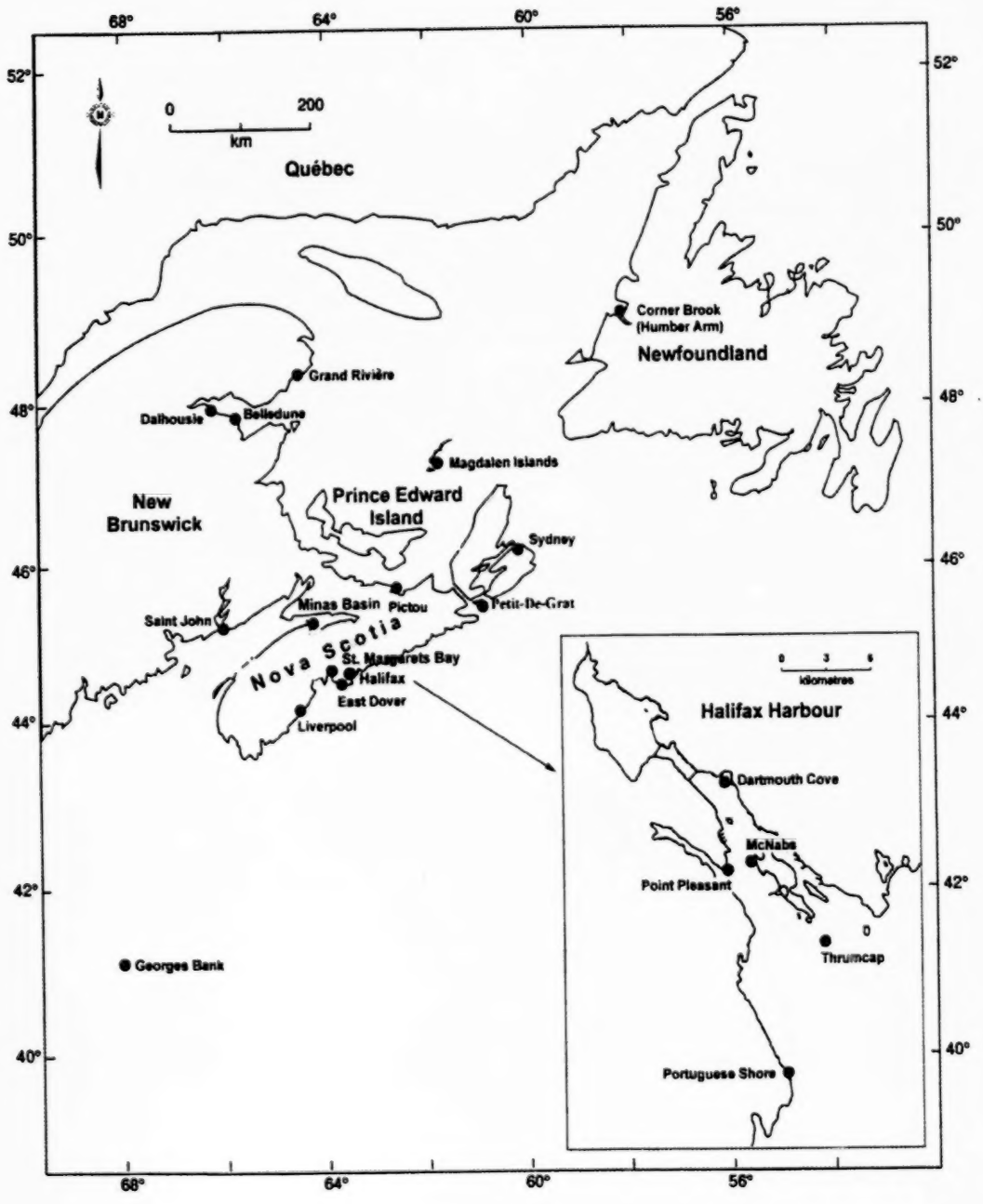

Figure 1. Lobster sampling locations